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A Hierarchical Treatment

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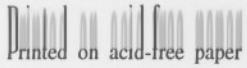

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CHAPTER 4

Evaluation of Organic Contaminant Exposure in Aquatic Organisms: The Significance of Bioconcentration and Bioaccumulation*

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I. INTRODUCTION

Bioaccumulation of compounds by an organism reflects its exposure to contaminants from various sources over time and represents the balance between the flux into the organism and the loss from the organism through processes such as biotransformation and elimination. Bioaccumulation is therefore an important direct link between the external contaminant concentrations in the sources and the potential effect of contaminants at various levels of biological structure and function. Bioaccumulated contaminants that attain sufficient concentrations at a receptor site for sufficient duration exert pharmacological and/or toxicological effects on the organism. Thus, the extent of bioaccumulation can be employed as a surrogate for the concentration at the receptor. This chapter will review our understanding of the accumulation of organic contaminants by aquatic organisms from water, sediment, and food, the factors that influence the accumulation processes, models for predicting accumulation, and the utility of estimating bioaccumulation for hazard assessment.

When aquatic organisms are only exposed to nonpolar contaminants via water, there is a strong relationship between the dose at the receptor and the concentration in the water. Thus, the paradigm developed for aquatic toxicology exposures is that the dose at the receptor is proportional to the dose in the organism which is, in turn, proportional to the concentration in the water. This paradigm allowed hazard evaluation based on concentrations in the external

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media and the development of relationships between compound physical/chemical properties and toxicity, e.g., the relationship between LC50 and the octanol:water partition coefficient (K_{ow}) (Ikemoto et al., 1992). The use of external environmental concentrations for risk assessment has been generally accepted in environmental and aquatic toxicology (Suter, 1993) and has even been applied to exposure in sediments (Long and Morgan, 1990; Di Toro et al., 1991; Neff et al., 1988). The use of external concentrations relies on one dominant source for exposure and fails to recognize the relative importance of multiple routes of exposure and the rates at which transfers between sources and biota occur.

Bioaccumulation of contaminants is temporal (kinetic) in nature and depends on the conditions under which the accumulation takes place (Landrum et al., 1992a; 1994a). The steady state condition (e.g., the balance between the contaminant fluxes of infusion and loss) represents the maximal accumulation that can be attained for a given set of exposure conditions. However, conditions can change rapidly enough that steady state may not be attained except under controlled situations. Early work in aqueous media used exposures that were sufficiently long with constant conditions of external factors, such as temperature, to establish steady state. The methodology was difficult, however, and a kinetic approach for estimating steady state was developed for aqueous exposures (Branson et al., 1975; Neely, 1979). Short-term kinetic measures permitted determination of steady state through calculations which yielded similar results to the longer exposures and established the utility of a kinetic approach. This approach also allowed evaluation of the net bioaccumulation at other than steady-state conditions. The continued fostering of this approach has allowed understanding of relationships among the contaminant concentration, processes in the external environment, and accumulation of contaminants by biota (Landrum et al., 1992a).

In the following sections, we will discuss factors that affect bioavailability of organic contaminants to biota in the aquatic environment (Table 1). Special emphasis will be given to the measurement and prediction of bioavailable contaminants, along with the problems that hinder the accurate prediction of bioavailabile contaminants in real-world aquatic environments.

II. AQUEOUS EXPOSURES

A. BIOCONCENTRATION OF CONTAMINANTS

Bioconcentration is the accumulation of freely dissolved contaminant in water by aquatic organisms through nondietary routes. Many of the factors affecting this process have been reviewed (Barron, 1990). In water-only exposures, the primary route of uptake in fish is across the gill epithelium, but depending on the compound and body size, 25 to 40% of the total body burden may penetrate across the skin (Saarikoski et al., 1986). This dermal absorption is a particularly important route of exposure for non-polar contaminants into organisms containing a chitinous skeleton (Landrum and Stubblefield, 1991).

Table 1 Factors Affecting Bioavailability of Organic Contaminants in Aquatic Systems

Factors affecting bioavailability in aqueous exposures	Factors affecting bioavailability in sediment exposures
Dissolved Organic Material (DOM):	Total organic carbon (TOC):
Concentration of DOM	Concentration of TOC
Molecular structure of DOM	TOC composition
pH of exposure media	Sediment particle-size distribution
pK_a of contaminant	Organism lipid content
Co-contaminants	Sorption/desorption from particles
Temperature	Contaminant physical/chemical characteristics:
Organism elimination processes	Octanol/water solubility
	Contaminant concentration
	DOM and colloid concentrations in interstitial water
	Organism feeding behavior and life history

In many cases, the toxicokinetic behavior of the contaminant in aquatic organisms can be described by a first-order, one-compartment model (Spacie and Hamelink, 1985). The degree of bioconcentration at steady state, represented by the bioconcentration factor (BCF), depends both on the rate of absorption and the rate of elimination:

$$\mathrm{BCF} = \frac{k_1}{k_2} = \frac{C_a}{C_w} \tag{1}$$

where k_1 is the uptake clearance (e.g., ml/g organism/h), k_2 is the elimination rate constant for the compound (e.g., 1/h), C_a is the concentration in the organism at steady state, and C_w is the concentration in the water at steady state. Using the freely dissolved water concentration and assuming no biotransformation, this BCF represents the relative solubility of the compound in water versus the organism's tissues. (The BCF can be reduced by biotransformation processes and active elimination.) The BCF as used in hazard assessment is an estimate of the maximum potential for contaminant accumulation in aquatic organisms and is estimated from the log K_{ow} for nonmetabolized, nonionic compounds in fish (Neely et al., 1974; Veith et al., 1979, 1980; Mackay, 1982; Oliver and Niimi, 1983), invertebrates (Connell, 1988), and macrophytes (Gobas et al., 1991). This model assumes that accumulation is a partitioning process between the water and the organism, with no physiological barriers to affect the accumulation process. Differences in the regression lines representing the relationship between log BCF and log K_{ow} among organisms may well be related to differences in lipid content and composition (see below). Further, the variability in the relationships of log BCF and log K_{ow} can be reduced by working with separate relations that are limited by compound class (Schüürmann and Klein, 1988).

Linear relationships between log BCF and log K_{ow} with slopes approaching unity (Veith et al., 1979, 1980) suggested that thermodynamics dominated the steady-state condition and that molecules were essentially at chemical equilibrium. Closer study of log BCF and log K_{ow} regressions show that over a broad

range of log K_{ow} (1 to 6), the BCF is described rather well, but outside this range or when the study compounds only cover a narrow range of log K_{ow}, the BCF may not be estimated accurately. This occurs because the majority of these log K_{ow}–log BCF regression models have been developed for specific classes of contaminants, e.g., halogenated hydrocarbons or polycyclic aromatic hydrocarbons (Veith et al., 1980; McElroy et al., 1989). The linear relation between log BCF and log K_{ow} breaks down for strongly hydrophobic compounds (log $K_{ow} > 6$; Sagiura et al., 1978; Bruggeman et al., 1984; Muir et al., 1985; Opperhuizen et al., 1985; Gobas et al., 1989a). The maximal observed value for log BCF seems to result with compounds having a log K_{ow} between 5 and 6. This effect is thought to be due to difficulty of the molecules to penetrate membranes because of diffusion and blood flow rate limitations (McKim et al., 1985; Gobas et al., 1986; Hayton and Barron, 1990), and because Log K_{ow} is a poor model for the partitioning between water and fish lipids (Opperhuizen et al., 1988; Ewald and Larsson, 1994).

As the data base for the log BCF–log K_{ow} model grows, more complex models are required to explain the bioaccumulation potential from water. Slow cavity formation in lipid membranes has been suggested to slow or limit accumulation of hydrophobic compounds and to cause variation between the octanol-water and membrane-water partition coefficients (Gobas et al., 1988b; Schüürmann and Klein, 1988). Therefore, adding a factor for the compound solubility in octanol improves ($S_{octanol}$) the log BCF–log K_{ow} relationship and accounts for some of the nonlinearity at the higher log K_{ow} values for compounds that are strongly nonideal in both octanol and lipid, e.g.,

$$\log \mathrm{BCF} = -1.13 + 1.02 \log K_{ow} + 0.84\, S_{\mathrm{octanol}} + 0.0004\,(\mathrm{mp} = 25)$$
$$(n = 36, \quad r = 0.95) \qquad (2)$$

where mp is the melting point of a solid in centigrade, and all liquid compounds are assumed to have a melting point of 25°C (Banerjee and Baughman, 1991). This equation allows additional prediction of bioconcentration for large hydrophobic molecules, such as dyes, and for polar compounds. However, compounds such as octachloronaphthalene are still not well predicted, presumably because of limited gill penetration. All these models contain the same assumptions of negligible metabolism and ignore the importance of organism physiology.

Molecular size and shape of a hydrophobic compound can affect or even inhibit its accumulation. The determining factor appears to be molecular size or molecular volume rather than molecular weight. The inclusion of steric factors to describe the bioaccumulation of polychlorinated biphenyls in fish in the traditional log BCF–log K_{ow} relationship improved predictions of PCB accumulation (Shaw and Connell, 1984). Further, the structure of the phospholipid bilayer of the gill epithelium can restrict uptake of hydrophobic molecules of long chain length or large cross-sectional area (>9.5 Å) by

imposing a physical barrier to diffusion (Bruggeman et al., 1984; Opperhuizen et al., 1985). The toxicity of polydimethylsiloxane (PDMS), a large hydrophobic organic molecule, to different organisms is low (Hobbs et al., 1975; Aubert et al., 1985) because it does not significantly accumulate in fish or in benthic organisms, either through aqueous or dietary exposures (Opperhuizen et al., 1987; Kukkonen and Landrum, 1994a). The study of Saito et al. (1990) showed that, due to their large molecular size, the absorption of highly lipophilic macromolecules (log K_{ow} 14; size 2,000 to 50,000 daltons) was limited by low diffusion into gill membranes and resulted in no bioconcentration.

The role of stagnant water layers next to membranes during the uptake of various contaminants across fish gills has been studied (Gobas et al., 1986; Saarikoski et al., 1986). Two permeation processes, one membrane-controlled and one diffusion layer-controlled, are apparently responsible for the kinetics of accumulation processes (Gobas et al., 1986). The uptake rate of compounds under membrane control is proportional to the log K_{ow} of the contaminant, while for a diffusion layer-limited process, the uptake is independent of log K_{ow}. Such a two-level process control can explain the positive correlation between log K_{ow} and accumulation rates through fish gill for contaminants with log K_{ow} ranging from 1 to 4, but cannot explain the absence of correlation or even decreasing accumulation rate for contaminants with log K_{ow} higher than 4 (Pärt, 1989). Further, studies of the influence of log K_{ow} on the transport across the gill membrane suggest that the mechanisms controlling transport are nonspecific with respect to chemical structure. Additional factors, e.g., molecular volumes, molecular weight, and self-association, affect the transport of highly lipophilic compounds (McKim et al., 1985).

Despite these limitations, log K_{ow} remains an important parameter for estimating the BCF of compounds in aquatic organisms (Lyman et al., 1990). This relationship is dominated by the solubility of nonpolar compounds in the organism lipids; thus, organisms became viewed as bags of lipids floating in water, and BCF as a chemical equilibrium between the lipid and water. Further, the relationship between log BCF and log K_{ow} has led to the use of thermodynamic equilibrium models for describing the potential accumulation of nonpolar organic contaminants by aquatic organisms and has dominated the risk assessment of aquatic systems. This concept has dominated even when multiple sources and kinetic limitations are apparent.

B. FACTORS THAT INFLUENCE BIOAVAILABILITY IN AQUEOUS EXPOSURES

Only the bioavailable fraction of a compound in water can be accumulated by aquatic organisms (Hamelink and Spacie, 1977). Factors that may influence contaminant bioavailability include the exposure concentration and contaminant binding to dissolved organic matter or particles in water. The pH can also affect bioavailability of polar compounds through dissociation/association processes for acids and bases.

Dissolved organic material (DOM) affects the chemistry and ecology of aquatic habitats and, more importantly, the fate of pollutants. DOM can bind several types of hydrophobic organic contaminants in aquatic environments (Ogner and Schnitzer, 1970; Hassett and Anderson, 1979; Gjessing and Bergling, 1981; Carlberg and Martinsen, 1982; Carter and Suffet, 1982; Landrum et al., 1984; Hassett and Milicic, 1985; Sithole and Guy, 1985; Chiou et al., 1986; Lara and Ernst, 1989; Lee and Farmer, 1989; Kango and Quinn, 1992). Some studies suggest that the binding occurs rapidly (McCarthy and Jimenez, 1985a; Schlautman and Morgan, 1993). In surface waters from different sources, the affinity of DOM for a given compound appears to differ (Carter and Suffet, 1982; Landrum et al., 1985; Morehead et al., 1986; Kukkonen and Oikari, 1991). The causes of these differences in binding affinity are not fully described. However, several phenomena have been postulated, and two or more binding interactions may occur simultaneously, depending on the chemical characteristics of the compound and the DOM (Choudhry, 1983). The interactions most likely involved are van der Waals forces; hydrophobic bonding, hydrogen bonding, charge transfer, ion exchange, and ligand exchange (Choudhry, 1983). The exact mechanism for a particular binding site will depend on both the chemical characteristics of the contaminant and the DOM.

The magnitude of contaminant-DOM binding is generally expressed as a partition coefficient, K_{oc} (e.g., low K_{oc} values represent less contaminant-DOM binding than high K_{oc} values), and for nonpolar organic contaminants is generally linearly related to K_{ow} as an expression of contaminant hydrophobicity (McCarthy and Jimenez, 1985a; Chiou et al., 1986; Lara and Ernst, 1989). The partition coefficients determined for various hydrophobic organics vary by factors of 5 to 10 among various soil and aquatic humic sources of dissolved/colloidal-bound forms in water (Carter and Suffet, 1982; Chiou et al., 1986). A reason for this variation in partitioning may be that contaminant binding is affected by the structure of the DOM. For example, pyrene binding to DOM in natural waters correlated with DOM aromaticity (Gauthier et al., 1987); an increase in aromaticity was shown to increase strength of PAH binding. Further, the binding of benzo(*a*)pyrene to DOM correlated with aromaticity, molecular size, and hydrophobic acid content of DOM (McCarthy et al., 1989; Kukkonen and Oikari, 1991). The structural features of DOM that influence binding vary among compound classes. For example, 2,2′,4,4′,5,5′-hexachlorobiphenyl will only bind strongly to the hydrophobic neutral fraction of DOM, while BaP will bind to both the neutral and the hydrophobic acid fractions. These examples demonstrate the complex interaction of compounds with DOM (Kukkonen et al., 1991).

The bioavailability of organic compounds in natural waters is decreased by DOM (Leversee et al., 1983; Carlberg et al., 1986; Kukkonen et al., 1989; Servos et al., 1989; Servos and Muir, 1989). The magnitude of the decrease is related to the magnitude of the contaminant-DOM partition coefficient, generally calculated as an organic carbon normalized partition coefficient (K_{oc}) (Landrum et al., 1985, 1987; McCarthy and Jimenez, 1985b; McCarthy et al., 1985; Black and McCarthy, 1968). An exception is naphthalene accumulation

from natural waters with a low DOM concentration (<4 mg carbon/l), which yielded two to three times higher BCF values than in DOM-free control water (Kukkonen and Oikari, 1991). A similar but less pronounced effect of DOM on naphthalene accumulation was detected by diluting a natural water sample to 5 mg carbon/l with DOM-free control water (Kukkonen et al., 1990). Likewise, the bioavailability of methylcholanthrene to *Daphnia magna* increased in water containing Aldrich humic acid (Leversee et al., 1983), but this could not be confirmed in another laboratory (McCarthy et al., 1985).

The observed bioaccumulation of model compounds in waters containing DOM can be compared to the predicted BCF values. The prediction is based on the assumption that contaminant bound to DOM is unavailable for uptake, mainly because the contaminant-DOM complex is too large to penetrate biomembranes and the dissociation rate is too slow to allow significant competition for uptake with the freely dissolved compound. Accordingly, bioaccumulation in water containing DOM is assumed to be proportional to the fraction of the contaminant that is freely dissolved. The present data on the effects of natural DOM on bioaccumulation suggest that correlation of freely dissolved contaminant with bioavailability is valid for short-term exposures, where little or none of the contaminant associated with DOM is available to aquatic animals, like *D. magna* (McCarthy et al., 1985; Kukkonen et al., 1989, 1990; Kukkonen and Oikari, 1991), *Diporeia* spp. (Landrum et al., 1985, 1987), *Crangonyx laurentianus* (Amphipoda) (Servos and Muir, 1989), and rainbow trout (Black and McCarthy, 1988). In addition to dissolved organic matter, the BCF of contaminants is reduced in systems where there is increased primary productivity. The ability of both dissolved and particulate organic matter to bind contaminants apparently reduces the amount that is bioavailable (Larsson et al., 1992; McCarthy and Black, 1988). The relative role of organic matter will depend on its binding capability (Eadie et al., 1990, 1992).

For ionizable compounds, the pH of the medium and the pK_a of the contaminant dictate the presence of nonionized and ionized forms. Nonionized forms are assumed to be the bioavailable forms according to the pH-partition hypothesis originally formulated for compound absorption in the intestine. Ionized forms are assumed not to penetrate the gut at significant rates (Klaassen, 1986). Consequently, for species exposed in an aqueous environment, the pH of the water strongly affects the toxicity and accumulation of organic weak acids, such as chlorophenols and dehydroabietic acid (McLeay et al., 1979a, 1979b; Saarikoski and Viluksela, 1981; Spehar et al., 1985; Fisher and Wadleigh, 1986; Saarikoski et al.,1986; Stehly and Hayton, 1990; Fisher, 1990, 1991). A similar potential is recognized for organic weak bases (Kalsch et al., 1991), although these compounds are not as well studied as weak acids.

The accumulation of pentachlorophenol (PCP) and dehydroabietic acid by *D. magna* and *Heptagenia fuscogrisea* is clearly pH dependent, both in artificial freshwater and in natural water containing aquatic DOM (Kukkonen, 1991). DOM reduced the uptake of PCP into *H. fuscogrisea* significantly when pH ranged from 4.5 to 7.5. However, at pH 3.5, the difference between humic and control waters was not statistically significant, although a 15% lower BCF

value was noted in humic water than in control water. At pH 8.5, there were no differences between humic and control treatments. Thus, the nonionized form of PCP is more readily accumulated by animals. This is also in line with other studies showing that the nonionized forms of compounds are more available and toxic than the ionized forms at lower pH (Saarikoski and Viluksela, 1981; 1982; Spehar et al., 1985; Fisher and Wadleigh, 1986; Fisher, 1990; Howe et al., 1994a).

The accumulation of weak organic acids cannot be fully described by the pH-partition hypothesis (Saarikoski et al., 1986; Pärt, 1989). Accumulation data indicate that the ionized compounds contribute significantly to the uptake rates and can affect toxicity at higher pH values (Stehly and Hayton, 1983). This effect happens when the exposure pH is well above the pK_a value (1 to 2 pH units) of the compound in question. There are three possible explanations for this phenomenon. The first considers the role of stagnant water layers at the gill surface. The diffusion rates in water of both nonionized and ionized forms are similar, and the diffusion resistance in the stagnant layers is independent of pH. For the nonionized forms with the highest K_{ow}, the diffusion through the stagnant layer will be rate-limiting. The uptake will proceed at the rate proportional to that of nonionized forms. When the pH increases above the pK_a, the relative importance of the stagnant layer gradually decreases, and the resistance in the membrane becomes rate-limiting (Saarikoski et al., 1986). The second possibility is that the gill membranes are permeable to the ionized form of contaminants. The third possibility is that the pH at the gill surface is different from that of the bulk water such that a portion of the ionized compound becomes nonionized.

The pH also affects the toxicity and accumulation of neutral compounds, although this effect is not as obvious as in the case of acids (Fisher, 1985; Fisher and Lohner, 1986). Accumulation of the nonpolar compounds, benzo(*a*)pyrene and lindane, by *H. fuscogrisea* and *D. magna* are also pH dependent; the highest BCF values are obtained at pH 6.5 and the lowest at 8.5 (Kukkonen, 1991). The pH-dependent accumulation pattern for lindane in these organisms is similar to that observed for the midge, *Chironomus riparius* (Fisher, 1985). The phenomenon is attributed to a lower permeation of lindane into the animals at pH 4 and an enhanced degradation of lindane at pH 8, compared to that at pH 6 (Fisher, 1985). Alternatively, the pH effect on the uptake rates of nonpolar compounds within the range from 5 to 10 has not always been observed (Pärt, 1989).

Other compounds in the water can influence the relative accumulation of a particular contaminant. When trace amounts of co-contaminants were added with particular contaminants to water, the bioavailability declined. For BaP, the more aromatic and hydrophobic the co-contaminants, (e.g., toluene) the lower the co-contaminant concentration that is required to reduce the uptake clearance (Landrum, 1983). However, water-soluble co-contaminants such as acetone do not exert any effect until they are in the 10% concentration range (Landrum, 1983). These effects are hypothesized to occur because of some

interaction between the BaP and solvent that produces a complex that is too large to penetrate the membrane or is slow to penetrate because of its size. In addition, solvent may disrupt membrane fluidity that may change membrane permeability to increase or decrease diffusion. However, the relative influence of co-contaminants has not always resulted in altered kinetics (McCarty et al., 1992).

In addition to organic matter and pH, temperature is an important environmental parameter that influences bioconcentration. Most exposures are conducted at constant temperature and the possible effect of temperature change is not known. That temperature has a profound effect both on observed toxicity and bioconcentration is known (Brown et al., 1967; Boryslawskyj et al., 1987; Landrum, 1988; Lohner and Fisher, 1990; Lydy et al., 1990; Lewis and Horning, 1991; Howe et al., 1994a). In general, there is a two- to fourfold (in some cases even tenfold) increase in toxicity with each 10°C rise in temperature (Fisher, 1991). This toxicity change corresponds with the concept that ectothermic metabolism increases approximately twofold for every 10°C increase. As the overall metabolism increases, the rate of uptake increases and the time required to reach a lethal body burden decreases (Gerould et al., 1983; Landrum, 1988). In some cases, the overall steady-state condition and the resulting effect remain constant, but the kinetics increase the time required to reach steady state at the lower temperature (Howe et al., 1994b). However, when biotransformation is important for determining contaminant disposition within an organism, increasing temperature may well increase the biotransformation rate, increase elimination, and decrease toxicity (Howe et al., 1994a; Niimi and Palazzo, 1985). Thus, the results of increasing temperature can enhance the rates of both the uptake processes and the biotransformation processes (Landrum, 1988; Lydy et al., 1990; Dabrowska and Fisher, 1993). There are, however, biological limits to such increases; at sufficiently elevated temperatures, organism tolerance will generally decline because its temperature tolerance is exceeded, e.g., anthracene biotransformation peaks at 25°C and declines at 30°C in chironomids (Gerould et al., 1983). Therefore, the influence of temperature on the overall uptake of contaminants can produce a confounding effect on the expected accumulation unless the temperature dependences on biotransformation, accumulation, and elimination processes are known.

In addition to uptake, elimination processes strongly affect contaminant concentration in the organism. Elimination occurs by two processes: (1) diffusion of the parent compound through the surface membranes of the organism (gills, skin), and (2) biotransformation and elimination of metabolites. In simple kinetic models, as described above, the mechanisms dictating the elimination are not always defined. Elimination is often measured as loss of radioactivity, which is loss of parent and nonparent metabolites. In rate constant terminology, this can be described as

$$k_e = k_p + k_m \quad (3)$$

where k_e is the overall elimination rate constant (1/h), k_p is the rate constant for physicochemical loss (elimination of parent compound, 1/h), and k_m is the biotransformation rate constant (1/h) (de Wolf et al., 1992). The relative contribution of the k_m to the k_e will depend on the k_p. In instances when a high log K_{ow} and a low k_p are observed, even a limited change in the ability of the organism to metabolize the contaminant can have a substantial effect on the overall elimination rate (k_e). This phenomenon was demonstrated with 2,8-dichlorodibenzo-*p*-dioxin (DCBP) kinetics in fish (Sijm and Opperhuizen, 1988). Two groups of goldfish were exposed to DCBP; one was pretreated with piperonyl butoxide, an inhibitor of the cytochrome P-450-dependent monooxygenases. The elimination rate constant of DBCP in the pretreated fish was significantly lower than that in the untreated fish. The calculated rate constant for metabolism, k_m, was significantly larger than the elimination rate constant, k_e, in the treated fish. However, in instances when a compound of low K_{ow} is observed along with a high k_p relative to the accumulation rate, the compound will have to be biotransformed at very high rates before the effect on the overall elimination rate can be noticed (de Wolf et al., 1992). Additionally, biotransformation can reduce the rate of total elimination in invertebrates (Landrum and Crosby, 1981). The idea of enhanced elimination, i.e., loss of parent compound plus metabolites, with biotransformation presumes a kidney-like function, where polar materials are filtered and cannot be reabsorbed. In invertebrates, however, this function does not exist, and biotransformation to more polar compounds may trap the metabolites in the organism unless other processes can actively transport the compound out of the organism. Such processes may include the formation of peritrophic membranes and fecal elimination.

C. NORMALIZATION OF CONTAMINANTS ACCUMULATED FROM AQUEOUS EXPOSURES

BCF is traditionally defined as contaminant concentration in the organism divided by the concentration in the water. However, recent work suggests normalizing contaminant concentration in the organism to the fraction of organism lipid. This will reduce the variance among organisms of differing lipid concentrations if the source is the same (Barron, 1990). However, variation in lipid composition or food chain position may still result in differences among species. When organisms collected from Lake Baikal were examined, the magnitude of the lipid-normalized BCF values were similar for two fish species, although the relationship between log $BCF_{\text{lipid normalized}}$ and log K_{ow} had significantly different slopes (Kucklick et al., 1994). However, when significantly different trophic levels were compared, seals had concentrations that were often ten times higher than fish, even after lipid normalization (Kucklick et al., 1994).

The BCFs for lake trout and white fish from Siskiwit Lake Isle Royal exhibit significant variability. Even with lipid normalization, the correlation of log K_{ow} with log BCF was weak for pesticides, while the correlation for PCBs

was even more variable (Swackhamer and Hites, 1988). More recently, the partitioning of 2,2′,4,4′-tetrachlorobiphenyl (TCB) between water and fish lipid appeared to be influenced by lipid composition (Ewald and Larsson, 1994). Fish lipid with high phospholipid content had essentially the same accumulation of TCB as fish of a low phospholipid content, but differences in lipid normalized BCFs were shown, suggesting species-specific accumulation. With the trend toward lipid normalization of organism data, care must be taken to ensure which method is being employed, since differences in the method can result in different lipid measures and therefore different normalized BCF values (Randall et al., 1991).

III. SEDIMENT EXPOSURES

Accurate prediction and evaluation of contaminant exposure and accumulation from sediments remains difficult because of the complex interactions between the contaminant, the sediment, and the organism. Factors that contribute to these interactions include:

1. Chemical characteristics and concentration of the contaminant
2. Physical and chemical characteristics of sediments
3. The presence of complex mixtures that can produce confounding effects when related to sediment constituents and biota
4. Organism behavior and physiology influenced by such environmental factors as temperature, nutrient availability, and habitat that can modify the exposure between species and temporally within a species
5. The length of sediment/contaminant contact time, which can change bioavailability

One approach to normalizing exposure of neutral organics against principal controlling factors in sediments uses an equilibrium partitioning bioaccumulation model (Lake et al., 1987; U.S. EPA, 1989). The basic hypothesis of this model is thermodynamic equilibrium. Thus, contaminant chemical activity is the same in each phase of a sediment matrix, including sediment particles, sediment carbon, interstitial water, and biota. For neutral organic compounds, the amount of organic carbon in the system generally determines the extent of contaminant partitioning between the sediment particles, interstitial water, and dissolved organic carbon. Thus, the exact route of contaminant exposure is not needed to determine the exposure or biological effect produced in the equilibrated system, because all phases will be at equilibrium and equal chemical potential (Di Toro et al., 1991). If a system is at equilibrium, equilibrium partitioning theory predicts that bioavailability should be directly proportional to contaminant chemical activity in a particular phase (i.e., interstitial water) and inversely proportional to the organic carbon content of the sediment, since organic carbon, to a large extent, controls sorption to sediment particles (U.S. EPA, 1989).

The equilibrium partitioning model can be directly applied for estimating bioaccumulation potential of sediment-associated neutral organic contaminants in benthic macroinvertebrates (McFarland, 1984; Lake et al., 1987). Assuming that organic carbon is the only sink for neutral organic contaminants, and that lipids are the only sink in organisms, the following model can be constructed (Lee, 1992):

$$\frac{Ctss}{L} = AF\left(\frac{Cs}{TOC}\right) \tag{4}$$

where Ctss = tissue concentration at steady state (μg/g organism), L = fraction of lipids in organism (g lipid/g organism), AF = accumulation factor (g carbon/g lipid), Cs = concentration of contaminant in the sediment (μg/g dry weight), and TOC = mass fraction of organic carbon in sediment (g carbon/g sediment). According to this model, the derived AF should not vary among neutral organic contaminants, because partitioning is not a function of lipid or carbon composition (Gobas et al., 1989b). Using empirical relationships between log K_{ow} and log BCF, and log K_{ow} and log K_{oc}, the AF was estimated as 1.7 (McFarland and Clarke, 1989). Accumulation factors less than this value would indicate less partitioning into lipids than predicted, while values greater than 1.7 would indicate a greater uptake of contaminant than can be explained by the model. In addition, if the systems are at equilibrium, AFs should be constant among species and among sediments, regardless of the amount of lipid contained in the species or the amount of organic carbon content contained in the sediment.

The validity of this approach has been tested primarily through the use of toxicity tests. A number of toxicity studies have accurately predicted exposure via equilibrium partitioning theory (Adams et al., 1984; Ziegenfuss et al., 1986; Nebeker et al., 1989; Swartz et al., 1990). Based on data derived from such studies, Swartz et al. (1990) and Di Toro et al. (1991) found correlations among organism survival, interstitial water concentrations, and organic carbon-normalized sediment concentrations for fluoranthene and kepone (Adams et al., 1984). Likewise, sediment toxicity conducted with *Hyalella azteca* in sediments dosed with DDT decreased with increasing carbon concentration (Nebeker et al., 1989). Equilibrium partitioning accurately predicted DDT toxicity which accounted for the reduced bioavailability in proportion to the amount of organic carbon. However, *H. azteca* exposed to endrin-spiked sediments under the same test conditions demonstrated an increase in toxicity with sediment organic carbon (Nebeker et al., 1989). Under the equilibrium partitioning approach, the extent of toxicity should be explained by the normalization to TOC in the sediment, and should be valid for contaminants with both high and low K_{ow} values. While this held true for DDT, it did not for endrin. Equilibrium partitioning should have predicted toxicity more accurately for endrin (log K_{ow} 3.23) than DDT (log K_{ow} = 6.6), since sorption capacity of sediment should have been much less for endrin than DDT (Nebeker et al., 1989).

Although equilibrium partitioning theory assumes that, at equilibrium, all phases in a sediment matrix have the same potential for contributing contaminant to an organism, this assumption has been found to be an oversimplification. Sorption was a strong function of organic carbon content when toxicity was determined for a mixture of chlorinated ethers in sediment (Meyer et al., 1993). The toxicity to *H. azteca*, *C. tentans*, and *D. magna* depended on sediment organic carbon, strengthening the validity of the equilibrium partitioning approach. However, the interstitial water concentrations did not correlate with observed effects for the two sediment-dwelling species, *C. tentans* and *H. azteca.* Further, toxicity did not correlate with interstitial water concentrations, and LC50 values varied by up to a factor of six among sediments containing various amounts of organic carbon. For equilibrium partitioning, the exposure and, in turn, the bioavailability of a contaminant should be similar for all test species after normalizing for contaminant concentration differences in interstitial water and organic carbon content. However, the bioavailability of fluoranthene to three species (*H. azteca, D. magna,* and *C. tentans*) was not similar in three laboratory-dosed sediments possessing the similar physical and chemical characteristics (i. e., similar organic carbon, particle size fractions). Significant differences in species response were observed (Suedel et al., 1993), and three- to five-fold differences in EC50 values were seen among the three sediments.

The aforementioned equilibrium partition studies have used toxicity as a surrogate for exposure in contaminated sediments. In order for toxicity to serve as a surrogate, the contaminant must be toxic at concentrations within its aqueous solubility limit, assuming that the organism is at the same chemical activity as the aqueous phase (i.e., interstitial water). A more direct approach to validate equilibrium partitioning theory is to measure bioaccumulation of sediment-associated contaminants. Bioaccumulation assays are perhaps more appropriate for testing equilibrium partitioning, since, depending on exposure length, a steady state body burden can be obtained. Equilibrium partitioning assumptions were tested in two infaunal species, *Macoma nasuta* and *Nereis virens*, by examining accumulation of PCB congeners and fluoranthene from sediments that varied in composition and organic carbon content (Brannon et al., 1993). Although calculated AFs bracketed the theoretical value of 1.7, the range over which AFs varied among the sediments and among the individual species was approximately 600%. Variation in bioavailability for three sediment-ingesting species was sevenfold, based on accumulation values normalized to contaminant concentration and organic carbon (Harkey, 1993). A wide variation in calculated AFs was found for infaunal organisms exposed to a variety of neutral hydrophobic contaminants over a range of hydrophobicities (Lee, 1992). Mean AFs ranged from 0.1 to 10.9 and were inconsistent both among species and contaminants. For example, mean AFs calculated for total PCBs were as low as 0.4 and as high as 5.9, using the same indicator species, *Macoma nasuta* (Rubenstein et al., 1987; Ferraro et al., 1990a). For compounds of similar hydrophobicity, mean AFs ranged over an order of magnitude, as

with benzo(*a*)pyrene (mean AF = 0.3, Ferraro et al., 1990a) and chlordane (mean AF = 4.7, Lake et al., 1987). Overall mean AFs calculated for PCBs, DDD, and chlordane exceeded the 1.7 value by a range of 25 to 250% (Lee, 1992). These reports suggest that factors other than carbon and lipid content are responsible for the wide range of contaminant accumulation among sediment-dwelling species.

Where equilibrium partitioning theory fails to accurately predict exposure, one or more assumptions of the model have been violated. For significant differences between predicted and observed bioavailability, thermodynamic equilibrium may not have been achieved. Such would be the case where sediment is manipulated prior to sediment assays (i.e., using laboratory-dosed sediments for bioaccumulation studies). If equilibrium partitioning fails, then organisms exposed to environmentally resident contaminants, that we can assume to have reached equilibrium, should provide validation. Such field-validation studies are scarce. However, deviations from constant AF (Lake et al., 1990) and toxic response relative to interstitial water concentrations (Hoke et al., 1994) exhibit deviations with environmentally resident contaminants (e.g., historically contaminated sediments). The equilibrium partitioning approach tends to reduce the variance in toxicity response and bioavailability over the use of whole sediment concentrations, but is insufficient to predict bioavailability of hydrophobic organic contaminants closer than about a factor of ten. The method, however, does provide a point of departure for comparison with other approaches and an initial estimate of expected results.

Bioaccumulation varies with the characteristics of the contaminant. For oligochaete worms, the bioaccumulation factor for accumulation of chlorinated hydrocarbons from sediments was low and relatively stable between log K_{ow} of 3 and 4, increased at values between 4.5 and 6, and declined at log K_{ow} of greater than 6 (Oliver, 1987; Landrum et al., 1989; Figure 1). The variation in bioaccumulation with log K_{ow} does not appear to be complicated by exposures to mixtures. Contaminants in the environmental mixtures seem to behave independently when concentrations are at typical environmental levels (Oliver, 1987; Landrum et al., 1989; 1991). When the concentrations become extreme, then the chemistry will change and the bioavailability will change. One example is the influence of cosolvents on partitioning to suspended matter. Sorption coefficients decreased exponentially with increasing fractions of methanol-water and acetone-water cosolvents when soil solutions containing hydrophobic contaminants were evaluated (Nkedi-Kizza et al., 1985).

The predominant environmental factor affecting contaminant bioavailability is adsorption to particles and, for neutral hydrophobic organic compounds, the particle-associated organic matter. Yet it appears that normalizing accumulation data to the fraction of organic carbon in sediment is inadequate to account for all of the variability in the data. Partitioning studies indicate that contaminant sorption and bioavailability may be affected by different forms of organic carbon as well as by amount (Word et al., 1987; Suedel et al., 1993). For example, organic carbon composed of mineral forms such as coal may sorb

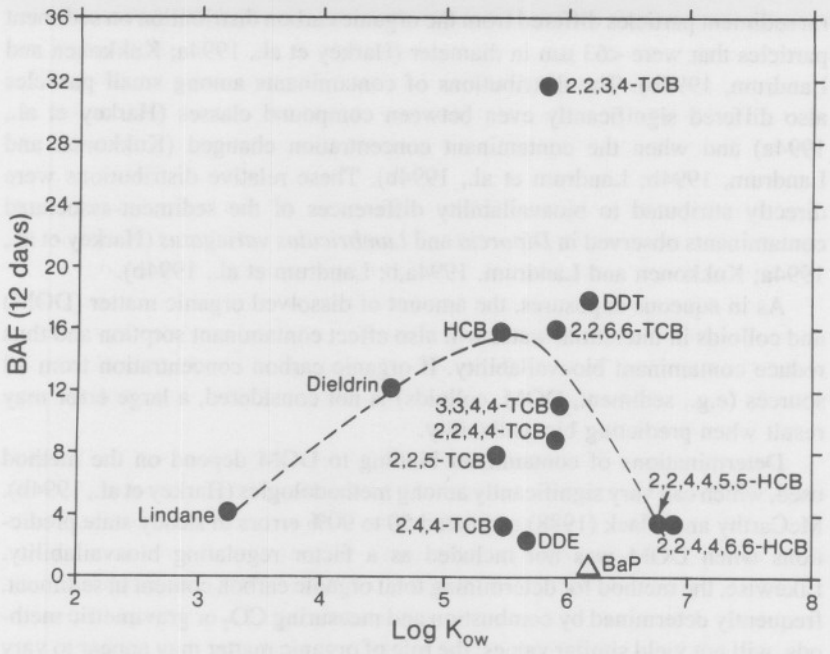

Figure 1 Scatter plot of the bioaccumulation factor (BAF), concentration in *Diporeia* spp. divided by the concentration in sediment, against log K_{ow}. The open triangle represents benzo[*a*]pyrene, the only compound that is not a chlorinated hydrocarbon. (Reprinted from Landrum et al., 1989. In: Aquatic Toxicology and Hazard Assessment XII, ASTM STP 1027, edited by U.M. Cowgill and L.R. Williams. American Society for Testing and Materials, Philadelphia, PA, pp. 315–329.)

hydrophobic contaminants more tightly or less tightly than dissolved or particulate forms of organic carbon (Suedel et al., 1993). The partitioning to soils was found to vary with soil organic matter composition. The organic carbon-normalized partition coefficient decreases for a particular compound with increases in the soil organic matter polarity expressed as the (O + N)/C ratio (Rutherford et al., 1992). The molecular structure as well as the amount of surface area comprising the various forms of organic carbon may largely control the bioavailability of neutral organics (Word et al., 1987). The differential bioavailability seen among sediments possessing the same organic carbon content but obtained from different sources (Word et al., 1987; DeWitt et al., 1992; Suedel et al., 1993) points heavily to the influence of sediment composition on exposure.

Not only does the partitioning apparently vary with the organic carbon content and composition, but compounds of different chemical classes appear to distribute differentially on various size classes of particulate material. Recently, the distribution of several nonpolar organic compounds among sediment particles was examined with respect to the organic matter content. In these studies, the distribution of pyrene, benzo(*a*)pyrene, and hexachlorobiphenyl

on sediment particles differed from the organic carbon distribution on sediment particles that were <63 μm in diameter (Harkey et al., 1994a; Kukkonen and Landrum, 1994b). The distributions of contaminants among small particles also differed significantly even between compound classes (Harkey et al., 1994a) and when the contaminant concentration changed (Kukkonen and Landrum, 1994b; Landrum et al., 1994b). These relative distributions were directly attributed to bioavailability differences of the sediment-associated contaminants observed in *Diporeia* and *Lumbriculus variegatus* (Harkey et al., 1994a; Kukkonen and Landrum, 1994a,b; Landrum et al., 1994b).

As in aqueous exposures, the amount of dissolved organic matter (DOM) and colloids in interstitial water will also affect contaminant sorption and thus reduce contaminant bioavailability. If organic carbon concentration from all sources (e.g., sediment, DOM, colloids) is not considered, a large error may result when predicting bioavailability.

Determinations of contaminant binding to DOM depend on the method used, which can vary significantly among methodologies (Harkey et al., 1994b). McCarthy and Black (1988) estimated 50 to 90% errors in steady state predictions when DOM was not included as a factor regulating bioavailability. Likewise, the method for determining total organic carbon content in sediment, frequently determined by combustion and measuring CO_2 or gravimetric methods, will not yield similar values; the role of organic matter may appear to vary if the differences in methods are not considered.

Partitioning between porewater and sediment particles has been described in multikinetic processes that appear as two differentially bioavailable pools: one in a reversible pool and another in a resistant pool (Landrum and Robbins, 1990). The fraction of contaminant that resides in each of these pools changes, depending upon the sorption duration, until equilibrium is reached. This may take months to years to achieve (Karikhoff, 1980; Di Toro et al., 1982; Coats and Elzerman, 1986; Witkowski et al., 1988; Fu et al., 1994). During this equilibration phase, contaminant bioavailability can also be expected to change with time, which can be especially important when examining data obtained from bioassays that employ spiked sediment. The duration of the mixing process affected toxicity in *Daphnia* sp., where toxicity decreased as the mixing time increased (Stemmer et al., 1990). Similarly, the bioavailability of phenanthrene and pyrene was found to change upon spiked sediment aging from 3 to 180 d; however, BaP exhibited no significant bioavailability changes, as determined by uptake rate coefficients (Landrum et al., 1992c; Harkey et al., 1994c). Differential bioavailability with sediment aging has also been observed for field-collected sediments. When *Macoma nasuta* were exposed to surface and deeper sediments (4 to 8 or 8 to 12 cm) of a sediment core, significant increases in calculated AFs were seen in surface sediments for 20 of the 29 exposures reported (Lee, 1991; Ferraro et al., 1990b). The higher AFs from the surface sediments were for contaminants that had spent less time in the sediment and were more bioavailable to the clams (Lee, 1991). However, the composition of the subsurface organic matter is most likely different from that at the surface. Subsurface material is likely more reduced due to diagenetic

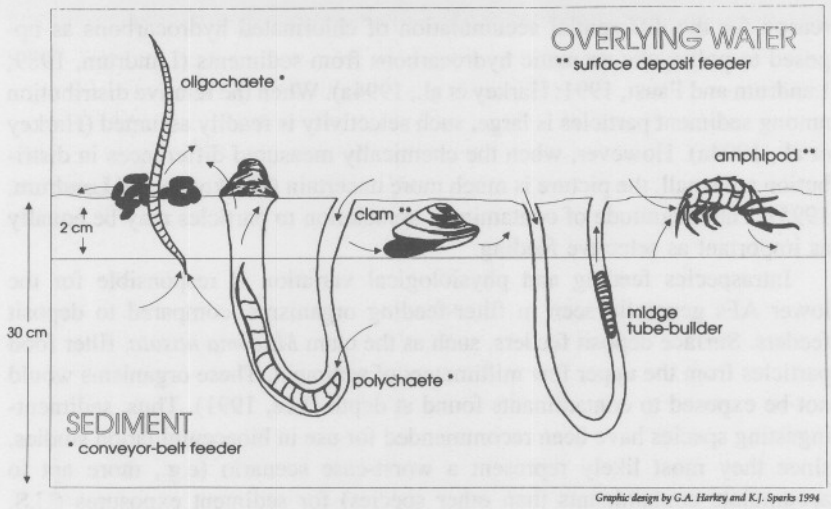

Figure 2 Representation of typical feeding behaviors of test species used in bioaccumulation assays. Surface deposit feeders ingest sediment from the sediment/water interface to the upper few centimeters, while conveyor-belt feeders and tube builders generally ingest sediment from deeper in the sediment core. Amphipods may occupy the sediment/water interface or burrow into the upper few centimeters of sediment, depending on species. Both oligochaete and polychaete worms ingest sediment from a variety of depths and deposit gut contents on the sediment surface so that uptake and elimination generally occur in surficial sediment. Arrows depict areas of contaminant uptake, elimination, and deposition by benthos via sediment particles and interstitial water. Biota are not drawn to scale.

(reconstructive) processes. As discussed above, changes in composition can influence contaminant bioavailability and may have contributed to the bioavailability differences observed with depth.

Epifaunal and infaunal sediment dwellers represent a myriad of feeding behaviors and life histories. Each of these behaviors can affect the relative contaminant exposure via manipulation of the environment surrounding the organisms (Figure 2). For example, infaunal oligochaetes burrow through sediment and obtain food from ingested sediment particles. These organisms, appropriately named "conveyor belt deposit-feeders," ingest sediment over a range of depths while they deposit gut contents on the sediment surface from posterior ends that protrude at the sediment-water interface (Karickhoff and Morris, 1985; Robbins, 1986). Bioturbation produced by these organisms disrupts any equilibrium established among sediment-associated contaminants, affecting bioavailability not only to the oligochaetes, but to all biota in the reworking zone. This behavior can also redistribute contaminants from buried deposits back into the feeding zone for shallower feeding organisms (Kielty et al., 1988a,b).

Selective feeding behavior is one of a number of biological characteristics affecting contaminant availability. In *Diporeia* spp., this selectivity combined with differential partitioning among sediment particles is suggested as a major

reason for the differential accumulation of chlorinated hydrocarbons as opposed to polycyclic aromatic hydrocarbons from sediments (Landrum, 1989; Landrum and Faust, 1991; Harkey et al., 1994a). When the relative distribution among sediment particles is large, such selectivity is readily assumed (Harkey et al., 1994a). However, when the chemically measured differences in distribution are small, the picture is much more uncertain (Kukkonen and Landrum, 1995). The magnitude of contaminant association to particles may be equally as important as selective feeding.

Intraspecies feeding and physiological variation is responsible for the lower AFs generally seen in filter-feeding organisms, compared to deposit feeders. Surface deposit feeders, such as the clam *Macoma nasuta,* filter food particles from the upper few millimeters of sediment. These organisms would not be exposed to contaminants found at depth (Lee, 1991). Thus, sediment-ingesting species have been recommended for use in bioaccumulation studies, since they most likely represent a worst-case scenario (e.g., more apt to accumulate contaminants than other species) for sediment exposures (U.S. EPA, 1989).

Physiological adjustments made among infaunal species, such as the production of tubes or burrows, can also alter the bioavailability of contaminants contained in interstitial water. These structures effectively encase the organism and create a differential permeability that tends to change the chemical composition of the water inside the tube compared to the surrounding interstitial water, so that direct contact with interstitial water does not occur (Lee, 1991; Aller, 1983; Aller et al., 1983). The resulting change in water flux through the burrow has been estimated to be between 2 and 2000 ml/h (Lee and Swartz, 1980; Figure 3).

In instances where contamination can be detected by organisms, sediment avoidance can occur, either by the animal burrowing around or under the zone of contamination (Landrum and Robbins, 1990) or by emerging to settle on the surface (Keilty et al., 1988c; Kukkonen and Landrum, 1994b). Of a more subtle nature, the exposure and accumulation of contaminants can result in contaminant-related changes in physiology. These changes in the physiology of the organism, which are manifest as changes in toxicokinetics, represent changes in accumulation and, in turn, bioavailability (Landrum et al., 1991, 1994b; Kukkonen and Landrum, 1994b). Both enhancement and reduction in bioaccumulation that occur with dose are thought to be strongly related to feeding rate for sediment-associated contaminants, because enhanced feeding rate can result in increased accumulation (Harkey et al., 1994a).

Once ingested by an organism, uptake of sorbed contaminants can be modified by gut processes that cause contaminant fugacity (apparent chemical activity of a chemical substance from a phase) and concentration to increase as the volume of food decreases and lipids are hydrolyzed (Lee, 1991; Gobas et al., 1988a, 1993). These digestive processes alter the thermodynamic gradient in such a way that there is a net flux of contaminant from gut contents into the gut wall that ultimately causes tissue contaminant concentrations to rise above what is predicted by equilibrium partitioning. Tissue concentrations of highly

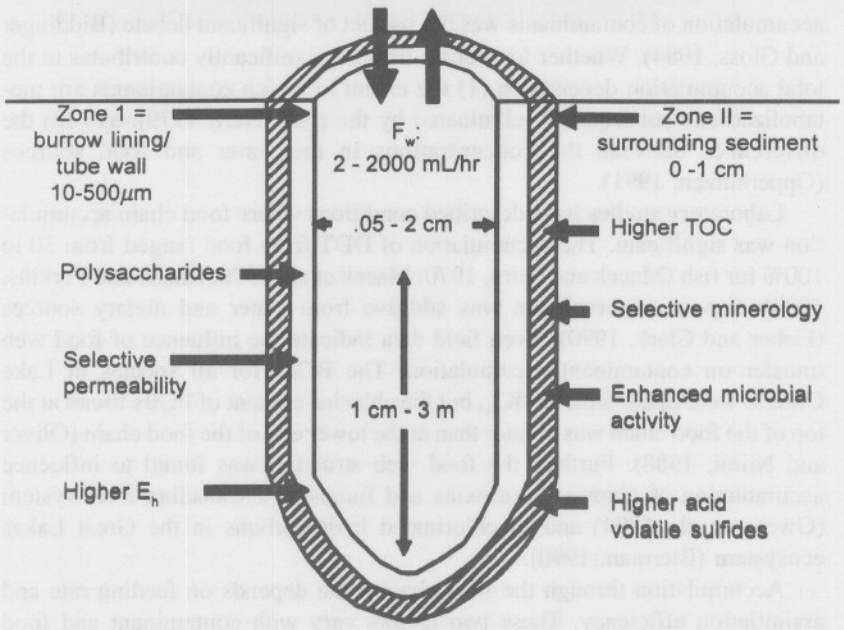

Figure 3 Properties of infaunal tubes and burrows. A generalized schematic to illustrate the structure and the range in the dimensions and flux of water (F_w or "irrigation") of tubes and burrows. The actual dimensions and shapes vary greatly among species. (Reprinted with permission from Lee, 1991. In: Organic Substances and Sediments in Water, Vol. III, Biological, edited by R. A. Baker, Lewis Publishers, Ann Arbor, MI, pp. 73–93.)

hydrophobic compounds have often been shown to exceed the concentrations predicted from thermodynamic partitioning (i.e., McFarland and Clarke's (1989) AF value of 1.7; Connolly and Pederson, 1988).

Other biological factors that may affect contaminant accumulation from sediments are organism age, sex, lipid content, and seasonality. Reproductive state and seasonal feeding behavior of species such as *Diporeia* affect accumulation and elimination of contaminants (Landrum, 1988). *Diporeia* spp. are particle-selective feeders that lack a continuous feeding pattern throughout the year (Quigley, 1988). As a result, their uptake and elimination kinetics are expected to differ from species that do not demonstrate a strong seasonal feeding behavior, e.g., oligochaetes.

IV. FOOD WEB TRANSFER

From various modeling efforts (i.e., the equilibrium model of organic chemical accumulation in aquatic food webs with sediment interaction; Thomann et al., 1992), it is apparent that identification of food web transfer and determination of assimilation efficiencies are major processes that require better description. The relative role of food chain transfer and biomagnification in the

accumulation of contaminants was the subject of significant debate (Biddinger and Gloss, 1984). Whether food chain transfer significantly contributes to the total accumulation depends on (1) the extent to which contaminants are metabolized and subsequently eliminated by the prey (Neff, 1979), and (2) the differences between the concentrations in the water and food sources (Opperhuizen, 1991).

Laboratory studies have described conditions where food chain accumulation was significant. The accumulation of DDT from food ranged from 30 to 100% for fish (Macek and Korn, 1970; Macek et al., 1970; Rhead and Perkins, 1984); kepone accumulation was additive from water and dietary sources (Fisher and Clark, 1990). Even field data indicate the influence of food web transfer on contaminant accumulation. The BCFs for all species in Lake Ontario were linear with log K_{ow}, but the chlorine content of PCBs found at the top of the food chain was greater than at the lower end of the food chain (Oliver and Niimi, 1988). Further, the food web structure was found to influence accumulation of chlorinated dioxins and furans in a Canadian river system (Owens et al., 1994) and of chlorinated hydrocarbons in the Great Lakes ecosystem (Bierman, 1990).

Accumulation through the food chain route depends on feeding rate and assimilation efficiency. These two factors vary with contaminant and food characteristics and also with contaminant concentration. As the feeding rate increases, assimilation efficiency declines, because the residence time in the gut declines (Klump et al., 1987; Opperhuizen and Schrap, 1988; Weston, 1990). Likewise, as contaminant concentration in the food particles increases, assimilation efficiency declines (Opperhuizen and Schrap, 1988). Further, assimilation efficiencies from ingested food change with organism age (Sijm et al., 1992).

Assimilation efficiencies for chlorinated organics in fish exhibit a nonlinear relationship with K_{ow}; for compounds with log K_{ow}s up to approximately 7, uptake efficiency is constant. However, for compounds with log K_{ow}s exceeding 7, efficiencies decline as the K_{ow} increases such that $1/E_0 = 5.3\ (\pm 1.5) \times 10^{-8}$ $K_{ow} + 2.3\ (\pm 0.3)$, where E_0 is the assimilation efficiency (Gobas et al., 1988a). These efficiencies for fish were measured with high quality (e.g., highly nutritional) food. The quality of the food can strongly affect the observed assimilation efficiency. With zebra mussels feeding on suspended sediment or algae, assimilation efficiencies for the suspended sediment particles were approximately 30% and, for algae, nearly 90% for hexachlorobiphenyl (Bruner, 1993). The mechanism for accumulation from the intestinal tract is apparently increased fugacity resulting from the assimilation of food materials (Gobas et al., 1993). This mechanism would account for the effects of feeding rate and food composition on assimilation. The increased feeding rate would reduce transit time as well as the fraction of food assimilated. Thus, the fugacity of the contaminant would not be increased as much as when a more complete assimilation of food occurs with slower gut passage. The difference in assimilation efficiencies can be explained by this same mechanism, where one food type is

better assimilated than another, e.g., algae versus sediment detritus. Thus, particle-selective feeding, coupled with differential contaminant partitioning to particles of differing composition, makes measurement of assimilation efficiencies for selective-feeding benthic organisms extremely difficult.

V. ESTIMATION METHODS

Because of the utility of log K_{ow} for predicting bioconcentration and partitioning to organic matter for nonpolar organic contaminants, methods that estimate log K_{ow} permit estimation of BCF. Beside direct measurement, empirical methods for estimating log K_{ow} include regression with aqueous solubility, partition coefficients with other solvents, or use of estimated activity coefficients (Lyman et al., 1990). Estimation of log K_{ow} from structure is best established through fragment analysis (Leo et al., 1971), and this method remains frequently used for log K_{ow} estimation. Other structural approaches have included the use of molecular volume (McGowan and Mellors, 1986), molecular connectivity (Kier and Hall, 1976; 1986), and more recently, linear solvation energy relationships (LSER, Kamlet et al., 1988). Among the methods that estimate log K_{ow} from structural properties of the molecules, the molecular volume method provides good estimates within a class of compounds. This is because the energy required for cavity formation within a solvent depends on molecular size, and the electronic character contributed by functional groups will remain relatively constant within a compound class. The molecular connectivity and LSER methods work well among a diverse mix of compounds, because both methods incorporate the change in molecular characteristics with changes in number and type of functional groups on the molecule.

The LSER model estimates a given property of a compound through multiple linear regressions of that property against several molecular parameters. The parameters are $V_I/100$ (intrinsic molecular volume divided by 100 to scale it to the other parameters), π^* (molecular polarizability), ß (the ability to donate a proton to a hydrogen bond), and α (the ability to accept a proton in a hydrogen bond) (Kamlet et al., 1986). The LSER model works best when a wide variety of molecular classes are included in the data set. This occurs because of the inclusion of dipole and hydrogen bonding characteristics of molecules in the model. Critical to the use of LSER is the determination of the parameters for each molecule. This leads to the following equation for estimating log K_{ow} (Kamlet et al., 1988):

$$\log K_{ow} = 0.35 + 5.35\left(\frac{V_I}{100}\right) - 1.044(\pi^* - 0.35\delta) - 0.35\beta + 0.10\alpha \quad (5)$$

($n = 245$, $r = 0.996$, sd = 0.131).

where δ is a modifier of the dipolarity (π*) to account for the influence of the aromatic nature of molecules and the influence of halogen atoms. A set of rules for parameter estimation has been published (Hickey and Passino-Reader, 1991).

Molecular connectivity is also very versatile. An additional feature is that the three-dimensional character of the molecule is implicitly encoded in some of the higher-level connectivity indices. The simple molecular connectivity indices are generated from the δ values, which represent the number of bonds to non-hydrogen atoms or the valence-corrected molecular connectivity that accounts for the number of valence electrons not participating in a bond to hydrogen. The level of connectivity can represent the environment of single atoms to multilevel connections of various path lengths, rings, or clusters within a molecule. The formal description of single-bond simple molecular connectivity, ^1X, would be calculated as follows for the single-bond paths in a molecule:

$$^1X = \Sigma(\delta_i \delta_j)^{-0.5} \tag{6}$$

where δ_i and δ_j are the number of non-hydrogen bonds for each atom (i and j) of a single bond path.

As the complexity of the calculation increases at higher levels of connection within a molecule, the use of computer programs to perform the calculations is suggested (L.H. Hall, Hall Associates Consulting, Quincy, MA). Following this approach, K_{ow} has been calculated for a wide range of nonionic compounds (Murry et al., 1978):

$$\text{(Hydrocarbons)} \qquad \log K_{OW} = 0.884\ (\pm 0.03)\,^1X + 0.41\ (\pm 0.09)$$

$$(r = 0.975,\ n = 45) \tag{7}$$

$$\text{(All other compounds)} \qquad \log K_{OW} = 0.95\ (\pm 0.01)\,^1X + 0.48\ (\pm 0.04)$$

$$(r = 0.986,\ n = 138) \tag{8}$$

Use of either LSER or molecular connectivity methods makes little difference for the purposes of determining log K_{ow}. In either case, to perform an estimate from a structure activity model, the parameters for the particular model need to be evaluated and the regression determined from a training set (a subset of the data representative of model parameters). In most cases, the greater the similarity of molecular structure in the training set to the compound of interest, the more likely the estimate will be accurate. Further, it is important to ensure that the conventions used for parameter estimation in the training set are consistent with those used for the compound of interest. There may well be

different assumptions employed by different researchers (i.e., whether to use or not to use a vector sum or sum with a component group of hierarchy of importance) to estimate the parameters, particularly for the LSER model (Hickey and Passino-Reader, 1991).

In addition to estimating log K_{ow}, both LSER (Park and Lee, 1993) and MC (Sabljic and Protic, 1982; Grovers et al., 1984; Connell and Schüürmann, 1988) can estimate BCF directly. For example:

Linear solvation energy relationships

$$\log BCF_{fish} = -0.95\ (\pm 0.23) + 4.74\ (\pm 0.25)\left(\frac{V_I}{100}\right) - 4.39\ (\pm 0.62)\beta + 0.88\ (\pm 0.38)\alpha$$

($r = 0.947$, $n = 51$; Park and Lee, 1993) (9)

Molecular connectivity

$$\log BCF_{fish} = -0.168\ (\pm 0.013)(^2X^V)^2 + 2.22\ (\pm 0.15)^2X^V - 2.32\ (\pm 0.38)$$

($r = 0.947$, SD = 0.3, $n = 20$; Sabljic and Protic, 1982) (10)

$$\log BCF_{fish} = 0.58^1X^V + 0.54$$

($r = 0.88$, $n = 49$; Connell and Schüürmann, 1988) (11)

where $^1X^v$ is the valence-corrected single-bond connectivity index.

These direct approaches for estimating BCF have the advantage that the log K_{ow} for the compound need not be known. Further, there is not the added variance of first estimating a log K_{ow} and subsequently a BCF. The BCF is estimated directly from compound characteristics and no measurements need be performed. When such estimates are made, the closer the compound characteristics are to the training set used to generate the structure-activity relationship, the more likely the estimate will be accurate. As with any such estimation, the variance of the correlation used for estimation must be considered when accepting the predicted value. Direct estimation of BCF from molecular characteristics is hindered because only a few equations are available. Such direct equations are currently available only for fish and for a narrow range of compounds, generally only polycyclic aromatic hydrocarbons and polychlorinated biphenyls. Because most of these regressions come from log relationships, the BCF estimates represent the medians and not the mean values when converted from log values to arithmetic values. If the mean BCFs are desired, then a correction for bias must be incorporated (Newman, 1993). Direct estimation that uses either LSER or molecular connectivity methods also helps overcome some of the complexities of nonlinearity in the log K_{ow} – log BCF relationship (Sabljic and Protic, 1988). The advantage of these methods lies in their incorporation of electronic and spatial characteristics of the molecule in

the relationship to estimate the BCF from the molecular structure. Disadvantages of these methods, however, ignore biological parameters, such as animal physiology and the role of biotransformation in determining contaminant accumulation.

VI. STEADY-STATE MODELS

The premise behind the use of equilibrium models for nonpolar organic contaminants is that accumulation of compounds is dominated by their relative solubility in the various phases, e.g., water, organism, and organic matter in other compartments (e.g., colloids, microparticulates). Equilibrium models, therefore, rely on the assumptions that (1) the compounds are not actively biotransformed or degraded, (2) there are no active (energy-requiring) processes dominating the distribution, (3) the conditions are stable enough that a quasi-equilibrium will occur, (4) environmental factors such as temperature do not change significantly to alter the equilibrium condition, and (5) organism and/or organic matter composition is not sufficiently variable to alter the distribution. For nonpolar organic contaminants, the solubility in organic phases as well as proportionality to K_{ow} has been widely recognized. This then led to development of relationships among various media for estimating bioaccumulation (Kenega and Goring, 1980) and has even been suggested as an approach for establishing sediment quality criteria (Di Toro et al., 1991).

The relative solubility of compounds among organic phases of the aquatic environment and in the lipids of organisms has led to development of equilibrium calculations between sediment and organisms. Because sediments represent the temporally integrated load to the aquatic system, the exposure of organisms should be better defined by relating bioaccumulation to the sediment concentration. These biota-sediment accumulation factors (BSAFs) are defined as the lipid-normalized concentration in organisms divided by the organic carbon-normalized concentrations in sediment (Ankley et al., 1992). These have been referred to in the literature as accumulation factors (AFs) while the inverse have been called preference factors (PF) (Landrum et al., 1992a; Lee, 1992). These BSAFs provide an estimate of accumulation from the sedimentary environment similar to the BCF from water. Such estimates were expected to reduce the variance compared to bioaccumulation factors (BAF, concentration in the organism divided by the concentration in the sediment or other reference compartment) which were typically reported on a wet weight of organism and dry weight of sediment basis. The first report that attempted to make this dual normalization suggested that the BSAF for nonpolar organic compounds would be a constant (McFarland, 1984). The utility of these concentration factors is limited due to the number of variables that affect accumulation from sediment (see above); the expected range for a given compound can exceed 100 (Lee, 1992). However, the relationship between sediment and bioaccumulation continues to be studied to describe the potential for ecological partitioning of compounds (Connor, 1984; Rowan and Rasmussen, 1992).

The use of thermodynamic equilibrium concepts to better estimate distribution among environmental compartments, and particularly to estimate accumulation in biota, was made simple and elegant through the use of the fugacity concept (Mackay, 1991). The fugacity approach even permits disequilibrium among compartments for modeling purposes so long as equilibrium within a compartment exists. When compounds can flow between compartments, the compounds will move down chemical activity gradients until the chemical activities (fugacities) are equal. This approach permits estimation of contaminants among environmental compartments either with simple equilibrium assumptions (level I) or nonsteady-state calculations (level III; Mackay, 1991). With level I calculations, the concentration in fish is estimated from the log K_{ow} – log BCF relationship; the concentration will depend on equilibrium distribution among the various phases. While the same relationship is employed at higher-level fugacity calculations, the concentration in the water that will dictate the BCF will be modified by various environmental processes prior to estimating the accumulation in fish (Mackay, 1991). Even processes such as biomagnification that allow the fugacity of fish to exceed that of water can be explained in terms of the enhanced fugacity in the intestinal tract of the fish feeding on various prey (Gobas et al., 1988a; Clark and Mackay, 1991). At the organism level, the fugacity approach can also accommodate more complex exposures and temporal changes to accommodate the complexities of the real world (Clark et al., 1990; Clark and Mackay, 1991). In all cases, the objective is to obtain a steady state estimate.

When multiple sources become significant for bioaccumulation, deviations from the expected equilibrium condition have been observed. The classic condition is food chain transfer and its relative importance for nonpolar persistent organic compounds. Even if the main source to the food web is water, the deviation from simple equilibrium becomes significant (Thomann, 1981, 1989; Thomann and Connolly, 1984; Connolly and Pedersen, 1988). Models that incorporate multiple contaminant sources must use some method of evaluating food chain transfer as well as accumulation from water. All these models demonstrate that the higher trophic level organisms have organism:water fugacity ratios greater than one. The elevated fugacity ratios demonstrate the incidence of biomagnification, i.e., the concentration in the organism relative to that of the water at steady state increases as the position in the food chain increases. Such biomagnification is apparently driven by the increased fugacity in the intestinal tract of organisms, as mentioned previously (Gobas et al., 1988a; Clark and Mackay, 1991). One absolute requirement for such biomagnification to take place is the inability of the organism to rapidly biotransform or eliminate the compound. The extent of biomagnification is also modified by the growth rate and alteration of the organism's lipid content. For example, increases in total lipid content decreased the elimination rate of polycyclic aromatic hydrocarbons (PAH) in *Diporeia* and *Hexagenia* (Landrum, 1988; Landrum and Poore, 1988).

Further, the recognized relationships between log K_{ow} and log BCF or partitioning to organic matter in sediments has allowed the development of

equilibrium partitioning models that, combined with food chain models, estimate the importance of sediments as contaminant contributors to the food chain (Connolly, 1991; Thomann et al., 1992). Equilibrium partitioning between sediment organic matter, interstitial water, and sediment-associated benthos is the basis of these models (Di Toro et al., 1991). These models also rely on relationships among bioaccumulation, lipid content, and the relative hydrophobicity of the contaminants. The various assumptions made in the equilibrium partitioning and food chain models permit the model predictions to bracket observed data. The fact that reasonable model results are achieved may be, in part, fortuitous. Issues such as growth dilution, which causes disequilibrium in species from fish to phytoplankton, accumulation, and uptake processes that are not directly coupled to respiration from water, are not considered. Alternatively, these pathways and processes may have little impact on the overall model. The models do indicate two areas that require significant development: (1) improved identification of the food chain links, and (2) improved data on assimilation efficiencies for ingested materials. Identification of food chain links can be improved through analysis of the stable isotope composition of food web members. As the trophic level increases, the stable isotope ratio increases, thus providing a quantitative approach to describing food web links. By better defining the food web in this way, models of food chain transfer can be made more reliable (Broman et al., 1992).

VII. USES AND LIMITS OF TOXICOKINETICS

Although equilibrium models give good approximations of contaminant distribution, kinetic models are needed to predict nonsteady-state, nonequilibrium accumulation from temporally and spatially varying exposures when the simplifying assumptions of the equilibrium partitioning models are inappropriate. This can be seen in the above examples, where either bioenergetics approaches (Connolly, 1991) or some form of food chain transfer was assumed that depended on estimated feeding rates and assimilation efficiencies (Thomann et al., 1992). The question becomes, can temporal variations be observed and is their magnitude sufficiently significant that kinetic models are warranted? For kinetics to be important in the bioaccumulation of contaminants, rates of the exogenous kinetic processes must be of similar magnitude as the biological processes, e.g., the rate of desorption or load is similar to respiration rate. The rates of importance for particular species will differ, because biological rates vary with organism size (Landrum et al., 1994a). For the amphipod, *Diporeia* spp., seasonal variation in the accumulation of PAH was observed and ranged from approximately a factor of 4 to 10 (Landrum et al., 1992b). This seasonal variation was best described by a kinetic model that incorporated both environmental and physiological variation in the kinetics (Landrum et al., 1992b). Likewise, the accumulation of PCBs by algae was seasonally dependent, where PCB uptake by algae was slow relative to growth (Swackhamer and Skoglund,

1991, 1993). Subsequently, thermodynamic equilibrium was not achieved. These nonequilibrium conditions limit the accuracy of the thermodynamic models described above, which often rely on equilibrium conditions, particularly at the lower end of the food web.

Kinetic models are particularly applicable for evaluating exposure from multiple sources and for examining changes in the exposure that may take place with either changes in concentration, chemical speciation, physiology, or environmental conditions. Kinetic models are also extremely useful for evaluating the important processes involved in contaminant bioaccumulation. Kinetic models can incorporate pathways that describe the mechanisms affecting the accumulation and loss of contaminants in organisms, and permit prediction of adverse biological responses using the tissue residue approach (Landrum et al., 1992a, 1994a). The disadvantage of such models is their need for relatively large amounts of data. Also, the kinetics for each species and the factors that affect the kinetics need to be known to parameterize the models. It then becomes a question of the need for time-dependent estimates versus the amount of effort required to achieve accurate models.

Kinetic models and their role in hazard assessment were recently reviewed (Landrum et al., 1992a). The formalisms that are available for compartment-based models include rate coefficient models, clearance volume models, and fugacity-based models. In the simplest cases, these models are mathematically equivalent. For the case of water-only exposure, the simplest rate coefficient model would conform to the following equation:

$$C_a = \frac{k_u C_w}{k_e}(1 - e^{-k_e t}) \tag{12}$$

where C_a is the concentration in the organism, k_u is the uptake clearance from water (ml/g organism/h), k_e is the elimination rate constant (1/h), C_w is the concentration in the water, and t is time (h). The uptake clearance in this and in other models is defined as the amount of source compartment scavenged of contaminant per mass of organism per unit of time. For this model, the concentration in the water is constant and there is no biotransformation. This model represents the simple uptake and loss of the parent compound such that at steady state, $BCF = C_a/C_w = k_u/k_e$.

In the clearance volume model, the BCF is equal to the volume of distribution (V_d). The V_d is the volume equivalent of a reference compartment, in this case water, that contains the same amount of compound as found in the organism. In this formulation, p is equivalent to k_u. This leads to the following integrated equation for accumulation:

$$C_a = V_d C_w \left(1 - e^{-\left(\frac{p}{V_d}\right)t}\right) \tag{13}$$

The argument for using this formalism suggests that k_e is an artificial value describing the fractional loss in concentration from an organism with time, which will depend on both physiological and environmental factors, while V_d is an equivalent referenced volume. In the case of k_e, changes in magnitude may result from both changes in the size of the storage compartment as well as changes in physiological function. Specific studies may well be required to determine which mechanism is applicable. With the above formulation of the clearance volume model, changes in V_d can also result from either of the mechanisms stated above. Additional data or a different model formulation may permit separation of the mechanism of importance.

When converting the above models to the fugacity formalism, the following equation examines the variability in the organism fugacity:

$$f_a = \frac{D_u f_w}{D_e}\left(1 - e^{-\frac{D_e t}{V_a Z_a}}\right) \tag{14}$$

where f_a is the fugacity in the organism (pascals), D_u is the uptake transfer coefficient (moles per hour per pascal), D_e is the elimination transfer coefficient (moles per hour per pascal), V_a is the volume of the organism (m^3), Z_a is the fugacity capacity of the organism (moles per cubic meter per pascal), f_w is the fugacity of the water (pascals), and t is time (h). Thus, at steady state, $D_u f_w = D_e f_a$ and the ratio of the fugacities $f_a/f_w = D_u/D_e$. If both sides of the equation are multiplied by Z_a/Z_w then $BCF = f_a Z_a/f_w Z_w = C_a/C_w = D_u Z_a/D_e Z_w$. From the above, the equivalency of the various formalisms is clear. However, as the complexity of the model increases, the equivalency becomes more difficult to depict; however, each of the approaches provides a good fit and accurate description of the experimental data (Landrum et al., 1992a). The selection of one model formalism over another depends on the experimentalist's experience and the ease of data collection. In general, the simplest model that will adequately address the question should minimize the errors associated with parameter estimation and, thus, result in the most precise estimates (Landrum et al., 1992a).

Despite the general utility of each of the above formalisms, the critical issue to the application of such models and their extensions to more complex conditions than water-only exposures is to ensure that all the assumptions of the model are explicitly described. For instance, in the above simple models, the water concentration (fugacity) must remain constant, and there should be no biotransformation. These two assumptions are easily recognized and are usually explicitly stated. What is less easily recognized is that the uptake clearances, rate constants, and transfer coefficients do not change over the course of the study. These assumptions are rarely recognized with the implications that go with them. For instance, if the coefficients change, then the integrated forms shown above are not correct. The fact is, these coefficients do change with the physiology of the organism and with a variety of environmental

factors. Thus, data used to generate such models are conditional, and the coefficients determined are conditional, based on the environmental and physiological conditions of the organisms under the conditions of the study.

To utilize a model that is useful for predicting accumulation in the field, the major factors affecting the coefficients for accumulation and loss must be identified. In the case of *Diporeia*, accumulation kinetics and the factors affecting kinetics were determined from a series of studies (Landrum, 1988,1989). These values and factors were combined into a seasonal model for field validation (Landrum et al., 1992b). Even when several of the factors that affect the coefficients were known, predictability was not accurate for the less hydrophobic compounds. Thus, the model still did not contain the appropriate coefficients needed to accurately estimate the accumulation of these less hydrophobic compounds.

Another rarely recognized assumption is that the systems must be homogeneous. If the organisms contact patches of contaminant, then the model assumption of a constant source would not hold. This may occur when organisms are initially added to sediments in typical sediment bioassays. It appears, particularly for some water-soluble compounds, that the initial accumulation is much faster than later in the experiment (Landrum, 1989). This was originally thought to be driven by sediment aging (Landrum et al., 1992c). However, more recent work with environmentally resident contaminants exhibits the same phenomenon of initial rapid accumulation followed by slower accumulation (Kukkonen et al., 1993). Accumulation of contaminants for organisms introduced into sediments initially occurs primarily from pore water, but the pore water is rapidly depleted of contaminant. Subsequently, accumulation occurs as a balance between pore water, replenished through contaminant desorption from particles, and assimilation of contaminant from ingested particles. The balance between which source dominates will depend on desorption kinetics for replenishing the pore water, the organism's selectivity, the organism's ingestion rate of particles, and the associated contaminant assimilation efficiency. Such nonhomogeneity invalidates the current models, i.e., models that assume homogeneity within a compartment, for sediment accumulation. New mathematical formalisms will need to be determined. The important point is that assumptions of the models need to be explicitly stated and care taken to ensure that there are no hidden assumptions for a particular application.

In addition to the classical compartment-based models described above, both bioenergetics and physiologically based pharmacokinetic models are other approaches for predicting concentrations in the organism and even concentrations in specific tissues (Landrum et al., 1992a). Bioenergetics models rely on the gross physiological functions of the organism, e.g., respiration and feeding, to provide encounter rates for the various source compartments. These physiological rates also are used to estimate the elimination and metabolism rates for compound removal and must be coupled to efficiency terms for compound transfer and transformation to produce the net compound accumulation or loss. Physiologically based pharmacokinetic models also include the details of transfer among organism tissues to produce concentrations in the

tissue containing the receptor. Either the compound concentrations or the fugacities among the various compartments can be used to track the compound in the two model approaches. For internal transfer among organs, the transfer coefficients between the circulating fluid, e.g., blood, and the tissues needs to be known. Both models can respond to the physiological state of the organism, so that as this changes, the net transfer to the organism can be estimated. In the aquatic environment, most of the models have been made for fish; in only a few cases (Boese et al., 1990) have these models been considered for lower organisms. The main advantage of these models lies in their strong tie to physiological function and their adaptability to organisms of various sizes, assuming that the routes of transfer do not change.

The assumptions in these physiologically based pharmacokinetic models are as important as in the compartment models. For example, it is important to understand the routes of accumulation for physiologically based models. With lower organisms such as amphipods, the accumulation from the aqueous phase may not be limited to the accumulation across the respiratory membrane. For *Diporeia*, the accumulation of nonpolar organic contaminants was found to exceed that of oxygen and depends on the surface area or surface area-to-volume ratio of the organism (Landrum and Stubblefield, 1991). Thus, to expand beyond simple equilibrium models for biomagnification, routes of transfer and loss that are often ignored in equilibrium models should be considered. The question of which model to use for predicting hazard is often difficult to answer, and a comparison of the various models needs to be thoroughly considered by the researcher (Table 2). The simpler the model used to fit the data, the more likely the accuracy of prediction under the same assumptions and conditions. It is important to use the models within the limits of their assumptions. As the model structure is changed, the inherent assumptions will likely change and must be evaluated and compared with the conditions under which the data are gathered.

VIII. BIOAVAILABILITY

Whether thermodynamic or kinetic models are employed, all the contaminant in a particular compartment may not be readily available for bioaccumulation. The fraction of material available in a compartment for uptake by biota is the bioavailable fraction. In water, the freely dissolved contaminant is generally accepted as the bioavailable fraction. No such simple definition can be applied for food or sediment sources.

The difficulty in applying the bioavailability concept is that first, a quantitative measure to define the bioavailability in a compartment needs to be made, and second, a compound that is temporarily unavailable can, after desorption or other change in chemical speciation, become available. One approach to addressing the first issue is a recent attempt to define the uptake efficiency as a measure of environmental bioavailability (Landrum et al., 1994a). Thus, environmental bioavailability (EBA) from a compartment can be defined:

Table 2 Comparison of Models Used in Exposure Assessment

	Model					
Model/attribute	**Equilibrium**	**Rate coefficient**	**Fugacity**	**Clearance volume**	**Physiological-based pharmacokinetic**	**Bioenergetic**
Requires assumption of equilibrium	Yes	No	No	No	No	No
Models multiple compartments	No	Yes	Yes	Yes	Yes	Yes
Models multiple uptake routes	No	Yes	Yes	No	Yes	Yes
Can be used to model internal distribution of toxicants	No	Yes	Yes	Yes	Yes	No
Potential to scale to other species	Yes (by lipid content)	Some	Some	No	Yes	Yes
Data requirements	Low	Moderate	Moderate-high	Moderate	High	High

Table modified from Landrum et al., 1992a.

$$\text{EBA} = \frac{\text{uptake clearance (ml source compartment/g organisms/h)}}{\text{encounter rate (volume/mass; ml/g/h)}} \quad (15)$$

The encounter mass for a solid phase may be substituted for encounter volume; the uptake clearance would then have units of g source/g organism/h, so that EBA would remain a fraction and reflect accumulation efficiency. While this approach presents a quantitative value that can be compared among different exposure conditions such as exposure in different sediments, the term is subject to both chemical and physiological factors that alter the uptake clearance. Further, measuring encounter mass or volume is difficult at best. Thus, EBA may not be easily determined in most situations. For intercomparison of a single organism and compound, comparative bioavailability can be described by comparing the uptake clearances from exposures to different sources, e.g., different sediments (Lee, 1991).

The second issue, temporal changes in bioavailability, from a risk assessment perspective will likely require that total contaminant entering a system be considered bioavailable. The issue is not whether a compound is bioavailable, but on what time scale it will be available. Because the prefix *bio* specifies an interaction with biota, the term bioavailable then will have temporal limits associated with the temporality of particular organisms. The time frames that are important change with organism size and behavior (Landrum et al., 1994a). While all of a compound entering a system may eventually become available for some species at some point in time, the focus for practical purposes must limit such discussions to time frames associated with a particular species of interest.

IX. UTILITY AND ASSESSMENT

The purpose behind estimating bioaccumulation is to determine the accumulated dose to a particular trophic level for modeling accumulation through the next trophic level, or to estimate the impact of that internal dose on the organism. The primary regulatory use of this type of data has been for human health risk assessment and not for assessing the impact either on the food chain or the effect of contaminants. Food chain models, as discussed earlier, often employ equilibrium partitioning approaches, particularly for the lower end of the food web. The use of more realistic kinetic models should help bring the data into line with that observed through field monitoring studies. They should also be useful for explaining seasonal variation, such as observed in the PCB accumulation in mussels with changes in lipid content (Capuzzo et al., 1989).

Evaluating the effect of contaminants on various levels of the aquatic food chain has traditionally used concentrations in the external environment. When multiple sources are involved and the exposure becomes complicated due to significant bioavailability limitations such as exposures in sediments, then assessing effects based on the external environment may not be very predictable.

Rather, there is a body of knowledge that is developing to evaluate the effect of chemicals based on the internal concentration in organisms (McCarty and Mackay, 1993). This is analogous to utilizing blood levels in mammals to predict drug effects and behavior. The complication in using this approach is that most researchers who have measured toxicity have not measured the organism's internal dose. Despite the lack of internal dose data for fish, concentration ranges exist for several mechanisms of action (McCarty and Mackay, 1993). These ranges lead directly to the application of data collected from bioaccumulation bioassays that are not usually used to assess environmental impact. Additionally, this approach is readily applied to mixtures and in the study of compound interactions (Landrum et al., 1989, 1991; McCarty and Mackay, 1993).

The range of concentrations producing effects varies with both mechanism of action and duration of exposure. Mortality due to narcosis, for instance, ranges from 2 to 8 mmol/kg for acute responses to 0.2 to 0.8 mmol/kg for chronic exposures in fish. Similar values for mortality have been observed for fish (McCarty and Mackay, 1993) and for selected invertebrates depending on the mechanism of action, e.g., narcosis by PAH in *Diporeia* (Landrum et al., 1991) and in *Daphnia magna* (Pawlisz and Peters, 1993a,b), decoupling of oxidative phosphorylation by pentachlorophenol in *Diporeia* and *Mysis relicta* (Landrum and Dupuis, 1990), and neurotoxicity by tri-*n*-butyltin in amphipods (Meador et al., 1991). The use of internal concentrations and their resultant effects has been applied to scope for growth in mussels (Widdows and Donkin, 1989). Thus, the approach may well be useful for other effects, although such applications remain to be revealed.

The ability to predict the accumulation of compounds will aid in the ability to predict effects. The total effect may not always depend on the total parent compound in the organism, but at some point the expected flux into the organism may need to serve as the dose for readily metabolized compounds. An alternative measure may incorporate the concentration of the metabolites for a specific chemical relating the effect to metabolite concentration, particularly for compounds that are activated by biotransformation, e.g., parathion to paraoxon. Thus, the use of internal dose may well take some refinement before it can be applied to a wide range of organisms and contaminant classes. The utility of the approach is, however, obvious. When organisms are exposed to multiple sources where none are dominant or where simple equilibrium models do not effectively reflect the concentration, then prediction of body burdens through kinetic models and assessment of effects based on internal dose should provide better estimates of environmental hazard.

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